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**PREDICTION OF THE THERMAL CONDUCTIVITY ANOMALY OF SIMPLE
SUBSTANCES IN THE CRITICAL REGION**

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PREDICTION OF THE THERMAL CONDUCTIVITY ANOMALY OF SIMPLE SUBSTANCES IN THE CRITICAL REGION

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SUMMARY

The anomalous behavior of the thermal conductivity of five gases, carbon dioxide, argon, nitrogen, oxygen, and methane has been investigated using the theoretical approach of Brokaw and the equation of state given by Bender. The analysis presented herein properly describes the trends in the data and in general is in good agreement with the data for $\rho/\rho_c < 1.1$. The theory falls 10 to 15 percent above the data at higher densities and appears about 10 percent high in the peripheral regions. The results seem adequate for engineering calculations and useful in establishing the thermal conductivity behavior of fluids for which no data have yet been taken.

NOMENCLATURE

A, B	constants in Eq. (15)
C_p	specific heat at constant pressure, J/gm-K
C_{p0}	specific heat at "zero" pressure, J/gm-K
$C_{pr} = C_p - C_{p0}$	"reacting" specific heat, J/gm-K
C_1	level adjustment constant
D_1	self diffusion coefficient, cm ² /sec
D_{1n}	binary diffusion coefficient, cm ² /sec
M	molecular weight, gm/gm-mole
n	number of monomers
P	pressure, MN/m ²
R	gas constant, J/gm-K
T	temperature, K
X^β	parameter defined by Eq. (12)
Y	parameter defined by Eq. (11)
Z	compressibility
λ	thermal conductivity, W/cm-sec
λ_R	contribution to thermal conductivity due to dissociation, W/cm-sec

λ_R^+	modified form of λ_R , W/cm-sec
λ_F	frozen thermal conductivity, W/cm-sec
λ_T^*	thermal conductivity at "zero" density, W/cm-sec
η	viscosity, gm/cm-sec
$f(\rho)$	a function of density, W/cm-sec
ρ	density, gm/cm ³
σ	diameter of the molecule, Angstroms
$\Omega(1,1)^*$	collision integral - self diffusion
$\Omega(2,2)^*$	collision integral - viscosity
X_1	monomer in the cluster
X_n	monomer
Subscripts:	
c	critical
pseudo	a representative value
CO ₂	carbon dioxide
N ₂	nitrogen
AR	argon
CH ₄	methane
O ₂	oxygen

INTRODUCTION

During the past decade, the debate on the existence of a "spike" in the thermal conductivity at the thermodynamic critical point has flourished. One of the earlier investigators to note this anomalous behavior was Guildner (1). Using a concentric cylinder cell, he found a marked increase in the thermal conductivity of carbon dioxide near its critical point. His conductivity data for selected isotherms, when plotted versus density, are nearly symmetrical about the critical density with some additional increase at the lower densities. Guildner (1) also suggested thermal conductivity to be extremely large if not of infinite value at the critical point.

Sengers (2, 3, 4) refined the experimental procedure. Using a parallel plate cell he acquired some excellent thermal conductivity data in the near critical region for carbon dioxide. He took great care to minimize cell convection and through careful analysis was able to show it to be negligible. In general, Sengers' peak conductivities are less than Guildner's for equivalent isotherms, perhaps indicating some undetected convection effects in Guildner's data. While some convection will probably be present in measurements using cylindrical cells, Bailey (5) took near critical argon data in both the horizontal and vertical positions without any noticeable gravitational effects.

Although the existence of the "spike" has been verified, procedures to predict this anomaly for various fluids are lacking. Herein we will combine the equation of state of Bender, Ref. 6, the theoretical approach of Brokaw, Ref. 7, the excellent carbon dioxide data of Sengers, Ref. 2, 3, 4, and other data to formulate a technique to predict the anomalous behavior of the thermal conductivity in the near critical region, for reduced densities in the range $0.6 < \rho/\rho_c < 1.5$.

It should be noted that Benders state equation Ref. 6, is analytic at the critical point. Thus in principle one should not expect to obtain a completely rigorous mathematical discussions of the anomaly which appears to be a mathematical singularity (2). However this does not seem to be of great importance for engineering applications.

THEORETICAL BACKGROUND

Brokaw's Analysis and Modifications

Brokaw, Ref. 7, assumed that the near critical thermal conductivity could be described in terms of a reacting gas. While he recognized that there exists a spectrum of cluster sizes, for simplicity, the gas was assumed to consist of a monomer and a single large cluster of n - monomers, i.e.

$$nX_1 \rightleftharpoons X_n \quad (1)$$

Based on the success of Brokaw's theoretical analysis in predicting the thermal conductivity of the $N_2O_4 \rightleftharpoons 2NO_2$ system, Ref. 8, this technique seems to be a sound approach. While the theory is rather involved, Brokaw represents the thermal conductivity as,

$$\lambda = \lambda_R + \lambda_F \quad (2)$$

where λ_R is the sum of the contribution due to dissociation and diffusion of the clusters, and λ_F is the rest; i.e. the thermal conductivity at "zero" density λ_T^* and the density effect contribution where

$$\lambda_F = \lambda_T^* + f(\rho) \quad (3)$$

is a function of density and λ_T^* depends only on temperature; the form of $f(\rho)$ was taken from Ref. 9 for the fluids considered.

The reacting contribution as defined herein is

$$\lambda_R^+ = C_1 \lambda_R = C_1 \rho C_{Pr} D_1 (D_{1n}/D_1) \quad (4)$$

The "reacting" specific heat C_{Pr} in Eq. (4) is defined as an excess specific heat

$$C_{Pr} = C_p(P, T) - C_{p0}(T) \quad (5)$$

and C_1 is a level adjustment constant. This constant is determined by matching the theory to a single experimental data point. Once C_1 is determined, the thermal conductivity values for the entire region can be found.

D_1 is the self diffusion coefficient and is given as

$$D_1 = \frac{6}{5} * 2.6693 \times 10^{-5} * \sqrt{T} / (\rho \sigma^2 \Omega(1,1)^*) \quad (6)$$

where

$$\frac{\rho D_1}{\eta} = \frac{6}{5} \frac{\Omega(2,2)^*}{\Omega(1,1)^*} \approx 1.32 \quad (7)$$

can be used to find D_1 . The ratio of the binary to self diffusion coefficient (D_{1n}/D_1) was argued by Brokaw, using scaling rules, to be of the form

$$\frac{D_{1n}}{D_1} = \left[\frac{2}{1 + (2.1)n^{1/4}} \right]^2 \left(\frac{1 + \frac{1}{n}}{Z} \right) \quad (8)$$

where the constant 2.1 is related to the geometry and density of the cluster and determined by Brokaw, Ref. 7, using $\partial P / \partial \rho|_T$ as determined by Sengers to give a good fit to the experimental carbon dioxide data of Refs. 2, 3, 4. Herein, it was found most expedient to retain the 2.1 value of Brokaw and adjust the level for each fluid through C_1 , (Eq. (4)).

The number of monomers in a cluster, n , was given by Brokaw and slightly modified herein:

$$n = \left\lfloor \frac{\left[\frac{RT}{\partial P} - 1 \right] / (1 - Z)}{\partial \rho} \right\rfloor \quad (9)$$

The absolute value was used to expedite computing in the peripheral regions when the symmetry constraint was not used. More properly, n should approach 1 and the reaction contribution approaches zero, in these regions.

SYMMETRY CONSTRAINT

We found by observation, that the data of Sengers, Refs. 2, 3, 4, possessed a high degree of symmetry about $\rho/\rho_c = 1$ for reduced densities in the range $0.6 < \rho/\rho_c < 1.5$. Guildner's earlier data (1) also reflected this symmetry. We, then, decided to neglect the slight asymmetry at lower density levels and for this prediction to define a pseudo value of density for $1. < \rho/\rho_c < 1.5$ as

$$\rho_{pseudo} = 2\rho_c - \rho \quad (10)$$

And since the computations are along an isotherm (isobar for argon) such a combination of (ρ_{pseudo} , T) gives a complete symmetry with respect to $\rho/\rho_c = 1$; thus Eq. (4) when evaluated at (ρ_{pseudo} , T) yields symmetric results. For example, the values of λ_F^+

at $\rho/\rho_c = 1.2$ with the symmetry constraint, Eq. (10), are identical to the values of λ_R^+ at $\rho/\rho_c = 0.8$ because

$$\left(\frac{\rho_{\text{pseudo}}}{\rho_c}, T\right) = (0.8, T)$$

SENGERS-KEYES ANALYSIS

Recently, Sengers and Keyes, Ref. 10, devised a scaling procedure for determining the near critical thermal conductivity of carbon dioxide. The essence of their work is illustrated in Fig. 1, taken from Ref. 10, where the data were correlated using the parametric grouping:

$$Y = \frac{(\lambda - \lambda_F) \sqrt{\rho/\rho_c}}{3.05 \times 10^{-5} \left| \frac{T - T_c}{T_c} \right|^{-0.6}} \quad (11)$$

$$X^B = \left(\frac{\left| \frac{T - T_c}{T_c} \right|}{\left| \frac{\rho - \rho_c}{\rho_c} \right|^{1/.35}} \right)^{.35} \quad (12)$$

Using Fig. 1 and Eqs. (11) and (12), one finds that for $X^B < 0.4$,

$$\frac{\lambda - \lambda_F}{3.05 \times 10^{-5}} \propto \frac{\sqrt{\rho/\rho_c}}{\left| \frac{\rho - \rho_c}{\rho_c} \right|^{1.71}} + f(\rho, T) \quad (13)$$

or thermal conductivity is a function of ρ alone.

For $X^B > 3$,

$$\frac{\lambda - \lambda_F}{3.05 \times 10^{-5}} = \left| \frac{T - T_c}{T_c} \right|^{-0.6} \sqrt{\rho_c/\rho} \quad (14)$$

It was found for $.4 \leq X^B \leq 3$, the data of Fig. 1 can be approximated (~ 3 percent error) by Eq. (15).

$$Y = \left[A + \frac{B}{(X^B)^{1/.35}} \right]^{(-.6)} \quad (15)$$

where $A \approx 1$ and $B \approx .9$. Note that the exponents represent those used to parametrize Y and X^B .

ANALYSIS AND RESULTS

Carbon Dioxide

The thermal conductivity of carbon dioxide has been carefully assessed by Sengers, Ref. 2, 3, 4. Fig. 2 illustrates the use of Bender's equation of state, Ref. 9, (for derived properties, see Ref. 11) and Brokaw's theoretical prediction of the anomalous

behavior, Ref. 7, for selected isotherms. The 305.23 K isotherm is compared to the data of Sengers, Refs. 2, 3, 4; as can be seen in Fig. 3, the data and theory agree quite well for $\rho/\rho_c < .85$ but the theory is about 18 percent high for $\rho/\rho_c \approx 1.4$. The important thing to note here is that while the prediction does not precisely match the data the general trends are in good agreement.

In order to obtain better agreement between the data and the theoretical prediction, the level was adjusted to fit the point at 305.23 K and $\rho/\rho_c = 0.8$. At the same time the symmetry condition, discussed in the Theoretical Section, was invoked and the constant C_1 in Eq. (4) was found to be 1.15, i.e.,

$$\lambda_{CO_2} = \lambda_F + 1.15 \lambda_R \quad (16)$$

In Fig. 3 the thermal conductivity for the 305.23 K isotherm, computed using Eq. (16), is again compared to the data of Sengers, Refs. 2, 3, 4. The results again indicate good agreement for $\rho/\rho_c < 1.1$, but the theory is about 10 percent high for $\rho/\rho_c \approx 1.4$.

In Fig. 4, the computed thermal conductivity using Eq. (16) can be compared to the data of Sengers, Refs. 2, 3, 4, for several selected isotherms. The data and theory are in good agreement for the 304.33 isotherm but the theory is about 13 percent high for the 313.13 K isotherm near $\rho/\rho_c \approx 1.4$. While agreement seems good near the critical point, further departure seems to indicate that the theory will overpredict the data by 10 to 15 percent. However, in the absence of data or as a guide to prediction trends, or in engineering use, the error is tolerable.

In Fig. 5, the thermal conductivity computed using Eqs. (11), (12), and Fig. 7 are compared to the data of Sengers Refs. 2, 3, 4, for isotherms. The computed values, which are in excellent agreement with that data as presented in Ref. 10, are herein 15 to 20 percent above the data, see Fig. 5. For $\rho/\rho_c = 1$ and the 304.33 K isotherm, the difference is about 20 percent. One must note however, that Bender Ref. 6, lists $\rho_c = 0.464 \text{ gm/cm}^3$ and $T_c = 304.21 \text{ K}$ for the critical constants while Senger (2,3,4,10) lists $\rho_c = .467 \text{ gm/cm}^3$ and $T_c = 304.19 \text{ K}$. Using Sengers critical constants instead of Benders lowers the predicted value by 8 percent. This illustrates the sensitivity of the results to selection of the critical constants.

Thus it appears that either technique, Keyes and Sengers (10) or Brokaw (7) will adequately describe the near-critical thermal conductivity of carbon-dioxide. The advantage of the technique of Ref. 10 is that it does not require a good equation of state in order to use the theory. The disadvantage is its extreme sensitivity to the selection of critical constants.

Nitrogen

With the fit to the carbon dioxide data as per Eq. (16), the question arises as to how well the "theory" will work for other fluids. In Ref. 12, Sengers reanalyzed the nitrogen data of Ziebland and Burton, Ref. 13, and based on two near critical data

†Due to a 0.02 K difference in critical temperatures of Refs. 2 and 6 the magnitude of the carbon dioxide isotherms of Ref. 2 were decreased by 0.02 K. This permits a more proper comparison between the data sources, Ref. 2 for thermal conductivity and Refs. 6 and 11 for PVT and derived properties.

points estimated the thermal conductivity curve for the 133.15 K isotherm. Using the theory of Brokaw, Ref. 7, as modified herein and Bender's equation of state, Ref. 6, the level was adjusted to fit the point as $\rho/\rho_c = 0.97$ and $T = 133.15$ K. The constant C_1 of Eq. (4) was found to be 1.35, i.e.,

$$\lambda_{N_2} = \lambda_F + 1.35 \lambda_R \quad (17)$$

The computation was then completed using Eq. (17) along the entire 133.15 isotherm and was found to match the other data point at $\rho/\rho_c \approx 1.49$. See Fig. 6. The estimated thermal conductivity curve given by Sengers, Ref. 12, and the computed curve are in good agreement except for $\rho/\rho_c \approx 0.6$ where the theory is about 15 percent high. Several other isotherms were computed using Eq. (4), both below and above the critical pressure. These results are illustrated in Fig. 6.

The unusual behavior of the predicted thermal conductivity beyond 1.5 ρ/ρ_c is believed to be due to errors in the derivative $\partial P/\partial \rho$ and an overextension of the theory.

Argon

In Refs. 10, 11, Bailey presented data for the thermal conductivity of Argon for several isobars, (as opposed to isotherms). The level was again adjusted to fit the point at $\rho/\rho_c \approx .65$ at 49.8 kg/cm² (4.8837 MN/m²) to determine the constant C_1 of Eq. (4). C_1 was found to be 1.2, i.e.,

$$\lambda_{AR} = \lambda_F + 1.2 \lambda_R \quad (18)$$

As can be seen in Fig. 7, there is a distinct shift in the data of Bailey, Refs. 5, 14, away from $\rho/\rho_c = 1$ to a peak near $\rho/\rho_c \approx 1.16$. This shift is unexplained. However there also does appear to be a small shift in the peak λ_{AR} as computed from Eq. (4) for the 50.8 kg/cm² (4.9817 MN/m²), see Fig. 7. Since the level predicted by the theory and the data are in good agreement in other regions, further modifications were deemed unwarranted in view of the unexplained shift in the thermal conductivity data toward higher ρ/ρ_c values.

The values of thermal conductivity were then calculated for several selected isotherms as shown in Fig. 8.

Methane

The methane thermal conductivity data of Sokolova and Golubev, Ref. 15, was most difficult to analyze. Moreover, the methane equation of state as given by Bender, Ref. 6, is probably the least well behaved, in the near-critical region, of the five gases correlated by Bender. In this case, the level was not adjusted to fit any particular point, but to best represent the trends of the 210 K and 216.3 K isotherms. As can be seen in Fig. 9, the 210 K isotherm data peaks near $\rho/\rho_c \approx 0.7$. There seems to be no apparent reason for this peak to occur as it does and is, as of now, unexplained. The 216.3 K isotherm data possess the more conventional appearance and are in better agreement with the trends of both the theory and other data, e.g. see Fig. 4. The constant C_1 of Eq. (4) was selected as 2.2.

$$\lambda_{CH_4} = \lambda_F + 2.2 \lambda_R \quad (19)$$

It is apparent from Fig. 9, that either the data of Ref. 15 for the 194.85 K isotherm are too high for the $\rho/\rho_c \approx 1$ region or the derived properties, Ref. 11, are too small. Also the adjusting constant (2.2) of Eq. (4) is much greater than for the other gases. As such it also seems out of line.

Assuming that Eq. (19) does represent the best fit to these data, the conductivity of methane can be computed. See Fig. 10. In Fig. 10, the dip in the 250 K isotherm can best be explained by the significant departure from the near critical region, i.e. an overextension of the theory, and troubles in the derived properties, Ref. 11. The λ_F or frozen thermal conductivity for the 250 K isotherm is included for comparison; it would seem that one is sufficiently far from the critical point such that Eq. (19) should no longer be applicable.

Oxygen

The equation of state for oxygen, Ref. 6, is quite well behaved and agrees very well with the results of Weber, Ref. 16, see also Ref. 11. As a result the theory should produce a reasonably good prediction of the thermal conductivity of oxygen. Assuming the level adjustment to be the same as for nitrogen,

$$\lambda_{O_2} = \lambda_F + 1.35 \lambda_R \quad (20)$$

the thermal conductivity of oxygen was computed for selected isotherms as illustrated in Fig. 11.

LEVEL ADJUSTMENT CONSTANT

While an a priori technique to predict the value of C_1 , Eq. (4), has not yet been established, there does appear to be a trend with molecular weight as seen in Fig. 12. This tendency to deviate with molecular weight may be related to geometry and density of the monomer which is in turn related to the constant in Eq. (8) and the number of monomer units (n), Eq. (9). If this trend is correct, a value of $C_1 = 1.3$ should be used for oxygen. In the absence of data, 1.35 was used. Note that methane does not appear to follow the trend established by the other gases.

CONCLUSIONS

The theoretical approach of Brokaw combined with an adequate equation of state as described by Bender can be used to predict the anomalous behavior of the thermal conductivity in the near critical region for five fluids: carbon dioxide, argon, nitrogen, oxygen, and methane. In general the theoretical prediction as described herein is 10 to 15 percent higher than the data for $\rho/\rho_c \approx 1.4$. The theory for $\rho/\rho_c < 1.1$ is generally in good agreement with the data. Methane is by far the most questionable of the theoretical predictions.

In this analysis the level adjustment constant, C_1 , is determined by matching the theory at a single data point. Once C_1 is determined, thermal conductivity values for the entire region can be found.

A trend has been noticed for the level adjusting constant with molecular weight. This effect is probably coupled with Brokaw's empirical determination of a constant which is related to the geometry, density,

and the number of monomer units.

The Keyes and Sengers technique also predicts the anomaly. The advantage of their technique is that it does not require an accurate equation of state; the disadvantage is that it is extremely sensitive to the choice of the critical constants.

It is felt that these approaches can be used to predict trends and give satisfactory engineering results.

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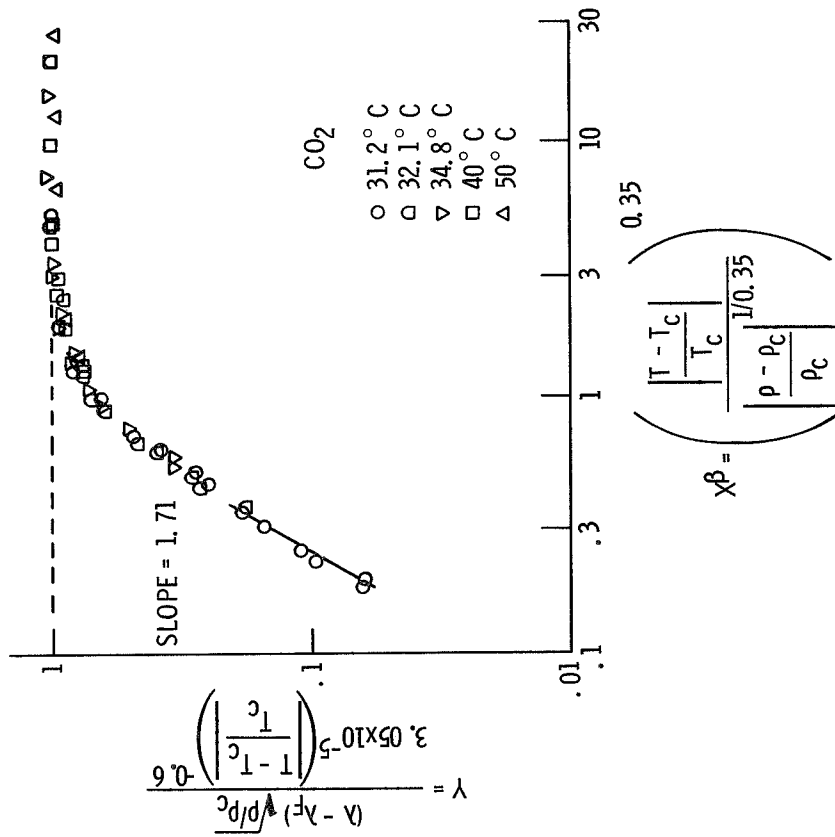


Figure 1. - Correlation of the thermal conductivity of carbon dioxide (taken from ref. 6).

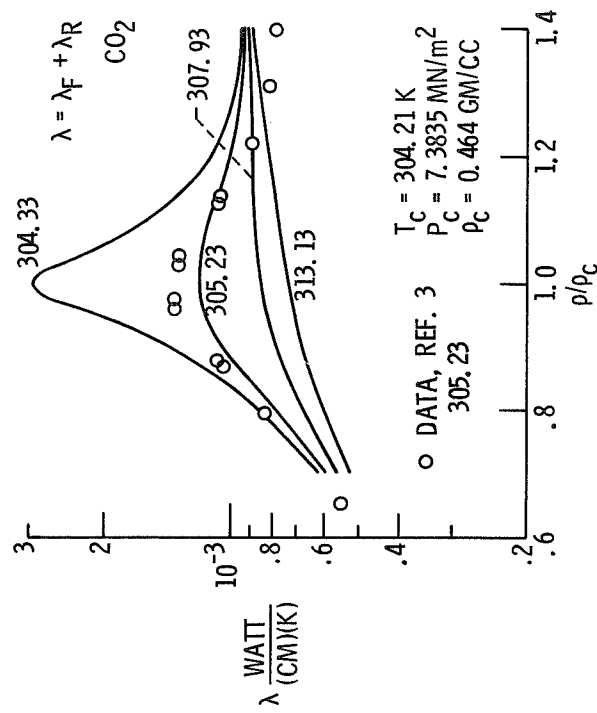


Figure 2. - Thermal conductivity of carbon dioxide, for selected isotherms, using Bender's equation of state (ref. 6) and the thermal conductivity analysis of Brokaw (ref. 7). Data from J. V. Sengers (refs. 2 to 4).

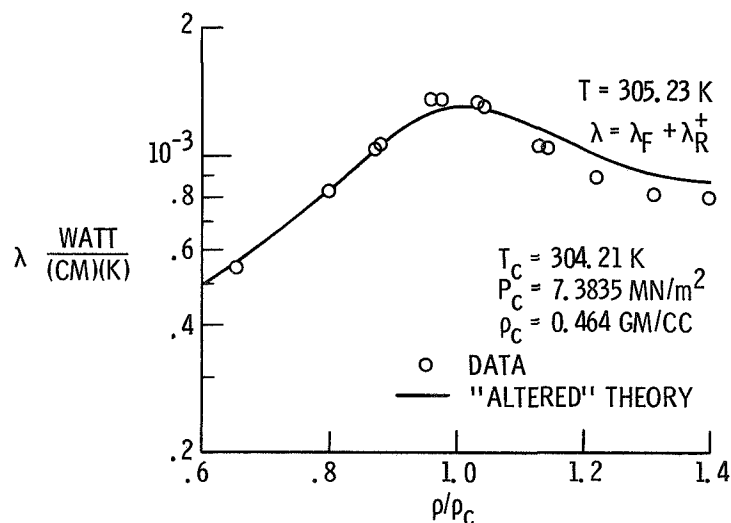


Figure 3. - Computed thermal conductivity of carbon dioxide adjusted for level at $\rho/\rho_c = 0.8$ and symmetry in λ_R , for the 305.23 K isotherm. Data from references 2 to 4.

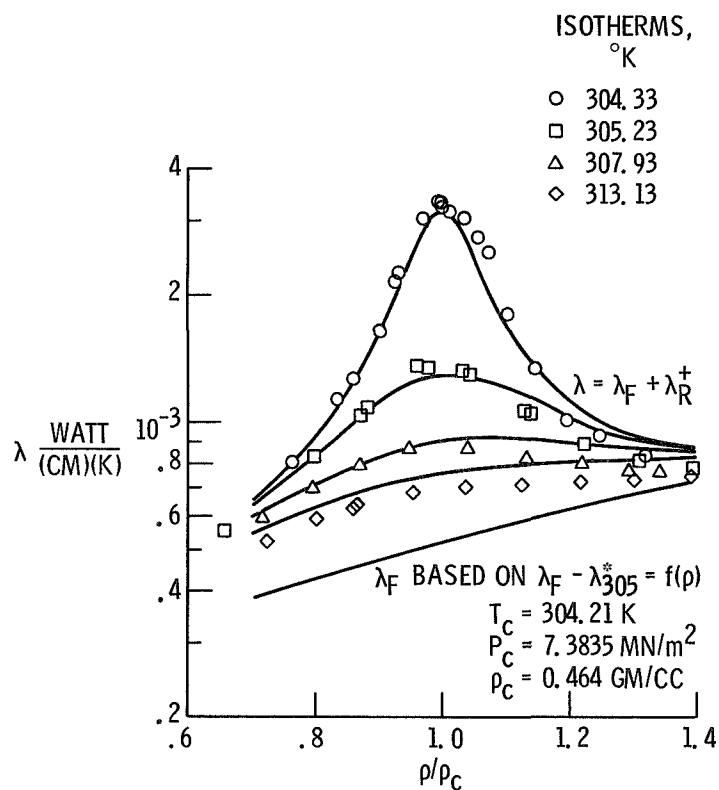


Figure 4. - Comparison of computed thermal conductivity of carbon dioxide with the data of Sengers (refs. 2 to 4) for selected isotherms. The "frozen" thermal conductivity is included for comparison (ref. 9).

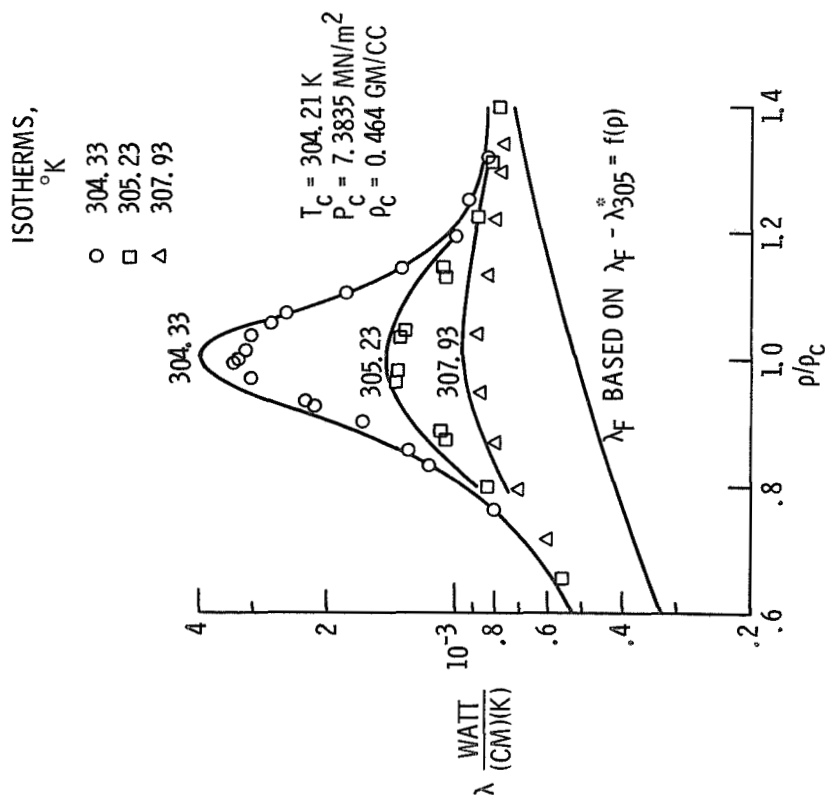


Figure 5. - Comparison of thermal conductivity computed by the Keyes-Sengers technique (ref. 10) with the carbon dioxide data of Sengers (refs. 2 to 4) for selected isotherms. The frozen conductivity is from reference 9.

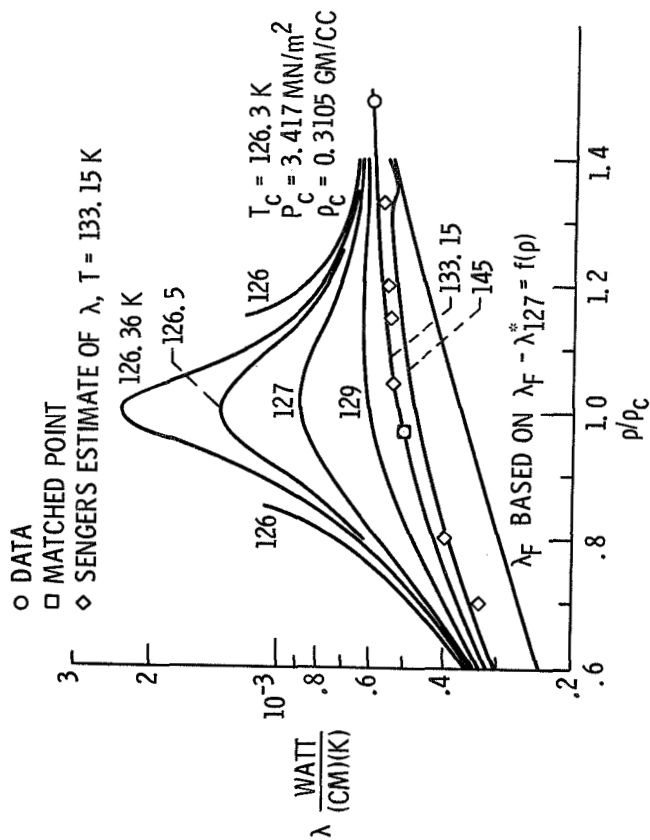


Figure 6. - Comparison of computed thermal conductivity of nitrogen with the estimated values from Sengers (ref. 12) at 133.15° K and other thermal conductivity values along selected isotherms.

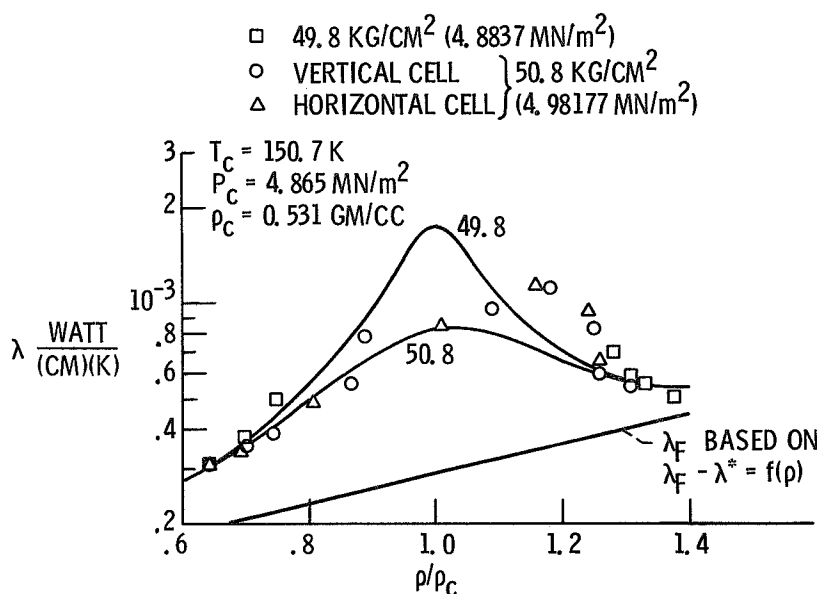


Figure 7. - Comparison of computed thermal conductivity of argon with the data of Bailey (refs. 5 and 14) for two isobars ($P_C = 48.392$ Bailey, $P_C = 48.014$ atm PROGRAM).

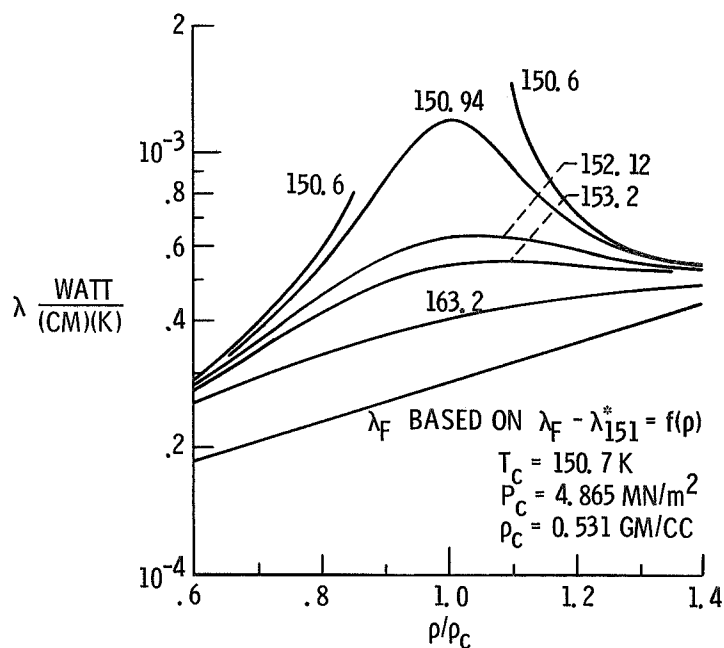


Figure 8. - Computed thermal conductivity for argon for a few selected isotherms.

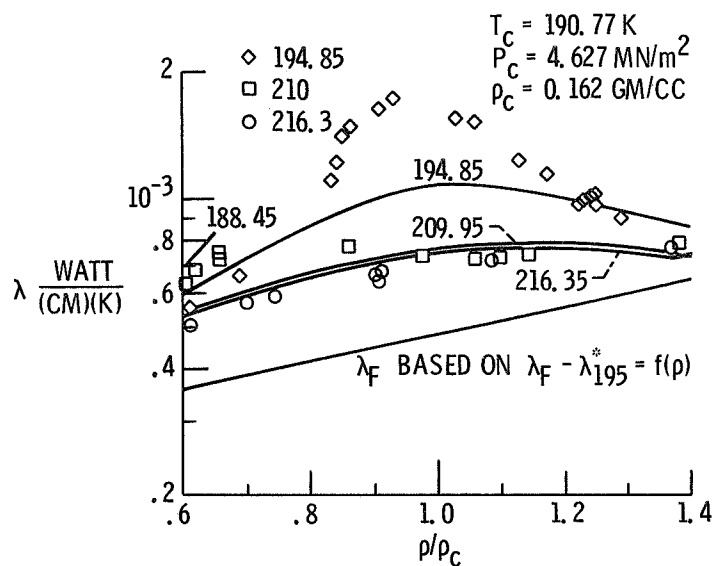


Figure 9. - Comparison of computed thermal conductivity of methane with the data of reference 15 for three isotherms.

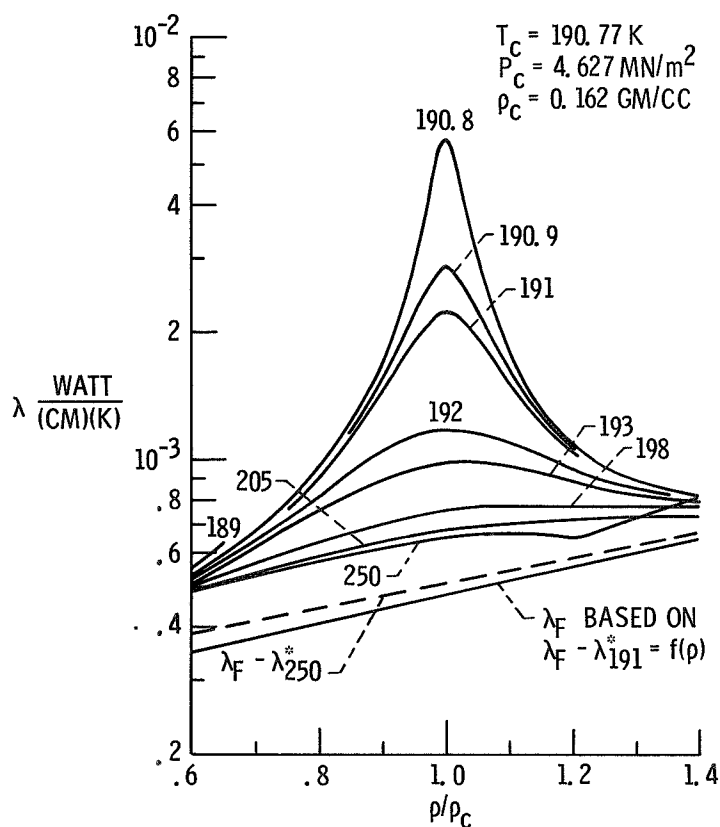


Figure 10. - Computed thermal conductivity for methane for selected isotherms.

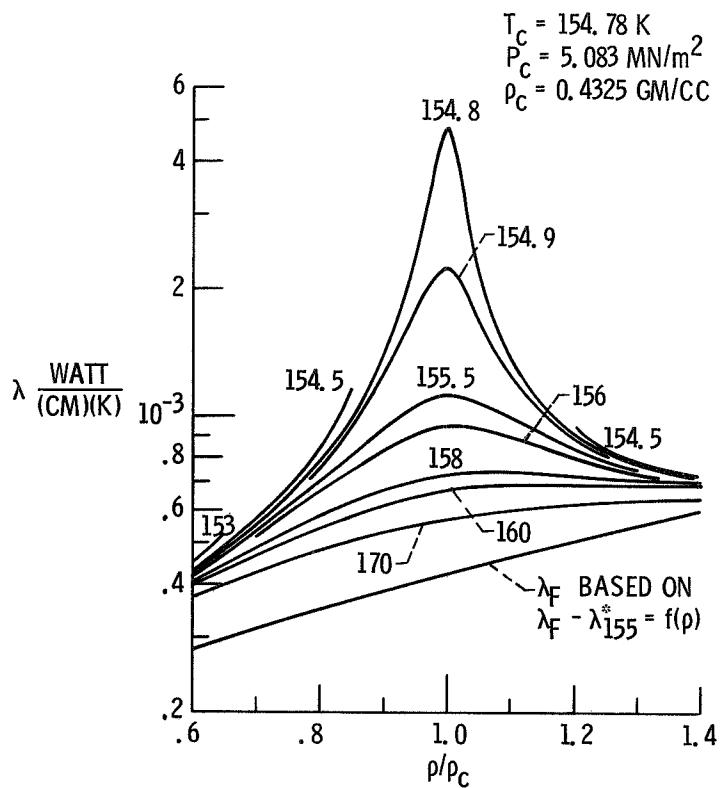


Figure 11. - Computed thermal conductivity of oxygen for selected isotherms.

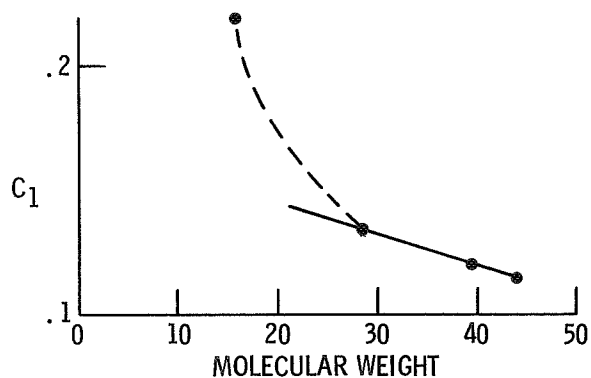


Figure 12. - Trend in the level adjustment constant C_1 with molecular weight.